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A SOLID-STATE, RECHARGEABLE, LONG CYCLE LIFE LITHIUM-AIR BATTERY (Postprint)

Binod Kumar, Jitendra Kumar, Robert Leese, Joseph P. Fellner, Stanley J. Rodrigues, and K.M. Abraham

Electrochemistry and Thermal Sciences Branch
Power Division

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14. ABSTRACT

This paper describes a totally solid-state, rechargeable, long cycle life lithium-oxygen battery cell. The cell is comprised of a Li metal anode, a highly Li-ion conductive solid electrolyte membrane laminate fabricated from glass-ceramic (GC) and polymer–ceramic materials, and a solid-state composite air cathode prepared from high surface area carbon and ionically conducting GC powder. The cell exhibited excellent thermal stability and rechargeability in the 30-105 °C temperature range. It was subjected to 40 charge-discharge cycles at current densities ranging from 0.05 to 0.25mA/cm2. The reversible charge/discharge voltage profile of the Li-O2 cell with low polarizations between the discharge and charge are remarkable for a displacement-type electrochemical cell reaction involving the reduction of oxygen to form lithium peroxide. The results represent a major contribution in the quest of an ultra high energy density electrochemical power source. We believe that the Li-O2 cell, when fully developed, could exceed specific energies of 1000 Wh/kg in practical configurations.

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A Solid-State, Rechargeable, Long Cycle Life Lithium–Air Battery

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This paper describes a totally solid-state, rechargeable, long cycle life lithium–oxygen battery cell. The cell is comprised of a Li metal anode, a highly Li-ion conductive solid electrolyte membrane laminate fabricated from glass–ceramic (GC) and polymer–ceramic materials, and a solid-state composite air cathode prepared from high surface area carbon and ionically conducting GC powder. The cell exhibited excellent thermal stability and rechargeability in the 30–105°C temperature range. It was subjected to 40 charge–discharge cycles at current densities ranging from 0.05 to 0.25 mA/cm^2. The reversible charge/discharge voltage profiles of the Li–O$_2$ cell with low polarizations between the discharge and charge are remarkable for a displacement-type electrochemical cell reaction involving the reduction of oxygen to form lithium peroxide. The results represent a major contribution in the quest of an ultrahigh energy density electrochemical power source. We believe that the Li–O$_2$ cell, when fully developed, could exceed specific energies of 1000 Wh/kg in practical configurations.
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The introduction of lithium ion batteries by Sony in 1991 has revolutionized the consumer electronics industry. The lithium ion battery business has grown into a multibillion dollar global industry, and a robust growth is anticipated in the future. The state-of-the-art lithium ion batteries employ many variations in cell components and chemistries. Most of them use a graphitic carbon anode (negative electrode), a liquid electrolyte comprised of lithium salts dissolved in organic solvents, a microporous polymer separator, and a lithium-intercalated transition-metal oxide cathode (positive electrode). The small lithium ion cells and battery packs made from them have gained worldwide consumer acceptance. Larger lithium ion batteries for hybrid and electric vehicles are yet to be developed, tested, and accepted. A few safety incidents and recalls of lithium ion cells produced by major manufacturers have also drawn significant attention.[1]

A high temperature environment can lead to the rupture, ignition, and even explosion of liquid electrolyte-based lithium batteries. For example, a lithium ion cell exposed to prolonged sunlight may rupture. A short-circuit in a lithium ion battery can cause it to ignite and explode. Some Li-ion cells are built with safety devices, which can prevent these hazards; however, separator defects and contaminants inside the cell can defeat those safety devices. The underlying causes for the safety concerns are the reactions involving liquid electrolytes, polymer separators, and transition-metal oxide cathodes. The societal needs in the present energy scenario require the development of inexpensive, thermally stable, and safe lithium batteries with high energy and power densities.

A need to develop new cell chemistries and batteries was articulated by Tarascon and Armand.[2] The importance of environmentally sustainable power sources was also recently highlighted in a review paper by Armand and Tarascon.[3] Theoretically, a Li–O$_2$ battery, an environmentally sustainable power source, is perhaps the highest energy density electrochemical power source that can be configured.[4] A Li–O$_2$ cell provides an open-circuit voltage (OCV) of around 3.0 V and a theoretical specific energy of 5200 Wh/kg if oxygen is contained in the battery. The oxygen need not be contained in the battery because it can be accessed from the air, and if such is the case, the theoretical specific energy of the Li–O$_2$ battery is 11,140 Wh/kg.[4] It is our estimate that a well-optimized Li–air

battery can yield a specific energy of up to 3000 Wh/kg, over a factor of 15 greater than the state-of-the-art lithium ion batteries. Indeed, the Li–O$_2$ cell may be the ultimate power source among electrochemical energy conversion and storage cells derived from lithium chemistry.

About 12 years ago, Abraham et al.[5] demonstrated the first practical nonaqueous rechargeable Li–O$_2$ battery cell. Their cell design employed a Li-ion conducting gel polymer electrolyte (membrane) derived from either a poly(acrylonitrile) or a poly(vinylidine difluoride) polymer. The cell exhibited good capacity utilization vs discharge rates, but its rechargeability was limited, attributed in part to the limited cycle life of the Li anode. Following this early investigation, several studies have been reported to improve the performance of the Li–O$_2$ battery. Abraham et al.[5] suggested the use of gel electrolytes with nonvolatile liquid plasticizers to increase battery life. Dobley et al.[6] studied the use of catalysts to enhance the rate capability and the capacity utilization of the oxygen electrode. Kuboki et al.[7] investigated ionic liquids in Li–O$_2$ cells to increase the overall stability and performance of the battery. Beattie et al.[8] investigated high capacity composite carbon cathodes to increase energy density. Ogasawara et al.[9] demonstrated the rechargeability of Li–O$_2$ cells starting with Li$_2$O$_2$ in the cathode with or without catalysts. Visco et al.[10] reported the electrochemical performance of a working Li–O$_2$ cell and suggested that practical Li–O$_2$ cells may lead to an energy density of 1000 Wh/kg or 1000 Wh/L. Despite these recent efforts, a rechargeable Li–O$_2$ cell with high energy efficiency and long life, in which charge and discharge proceed with low polarizations, has remained elusive. In this context, the attributes including excellent rechargeability of the Li–O$_2$ cell reported in this paper are believed to be a major contribution as they overcome the previously identified deficiencies.

In this paper, the electrochemical performance of totally solid-state, rechargeable Li–O$_2$ cells are presented and analyzed. In these cells, oxygen can be replaced by air from the atmosphere with modifications of the cell design. The primary component of the electrolyte and the cathode is a glass–ceramic (GC) material, which is very stable when in contact with water. A chemical degradation of either of the two components by the presence of water leading to cell failure is not anticipated.

Experimental

Materials and cell fabrication.— The polymer–ceramic (PC) membranes were prepared from dried poly(ethylene oxide) (PEO), LiN(SO$_2$CF$_2$CF$_3$)$_2$ (LiBETI) salt, Li$_2$O, and boron nitride (BN). In a

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Table I. Physical attributes of the components of Li–O$_2$ cells.

	Lithium anode		Membrane laminate			Cathode			
Li–O$_2$ cell	Thickness (cm)	Weight (gm)	Material	Thickness (cm)	Weight (gm)	Material	Thickness (cm)	Weight (gm)	Porosity (%)
1	0.038	0.040	PC(BN)/LAGP/PC(BN)	0.150	0.68	25% C* + 75% LAGP on Ni mesh	0.048	0.163	12
2	0.038	0.040	PC(Li$_2$O)/LAGP/PC(BN)	0.205	0.58	50% C* + 75% LAGP on Ni foam	0.045	0.172	27
3	0.038	0.040	PC(Li$_2$O)/LAGP/PC(BN)	0.101	0.46	25% C* + 75% LAGP on Ni foam	0.031	0.146	24
4	0.038	0.040	PC(Li$_2$O)/LAGP/PC(BN)	0.052	0.28	25% C* + 75% LAGP on Ni foam	0.058	0.164	24

C* is 60% PWA activated carbon + 40% Ketjen carbon black.

typical preparation, a dried mixture of the electrolyte components was energy milled and hot-pressed to obtain 200–400 µm thick membranes. The GC membrane was prepared from a batch of various oxides corresponding to the 18.5Li$_2$O:6.07Al$_2$O$_3$:37.05GeO$_2$:37.05P$_2$O$_5$ (LAGP) molar composition. The batch was melted at 1350°C in a platinum crucible, cast into solid glass sheets, powdered and pressed into a membrane, and sintered at about 850°C. The sintering parameters (temperature and time) were optimized to achieve the highest membrane conductivity. The conductivities of both the GC and PC membranes were extensively characterized, and the data have been reported in prior publications.[11-16] The cathode was prepared using nickel mesh or foam, carbon black (acetylene 50% compressed), Teflon (TE-3859), and GC powder. Nickel foam was used with an intent to achieve further enhancement in cell capacity. Carbon black (25 and 50 wt %), LAGP (50 and 75 wt %) powders, and Teflon were mixed with deionized water to obtain a soft mass, which was spread on both sides of the nickel mesh or foam and sandwiched between two stainless steel plates. The LAGP material used in the cathode is similar in chemistry and crystal structure to the LAGP material used in the membrane laminate. The assembled sandwich was pressed, dried at 100°C overnight, and sintered at 240°C for 20 min in an argon atmosphere. The setup of the cells consists of metallic lithium as the anode, three membrane laminates (two PC layers and one LAGP layer), and a cathode. The cell area in all cases was 2.00 cm^2. The cells were assembled in a dry box (O$_2$ < 80 ppm, H$_2$O < 89 ppm) contained in the casing with a spacer and an O-ring. The cathode side of the casing was perforated to allow the flow of oxygen to the cathode. The cell components along the casing were pressed to seal using a Hohsen cell maker (Hohsen Corporation, 10-4-601, Minamisenba, 4-Chome, Chuo-ku, Osaka, Japan) into a working cell.

Electrochemical characterization.— The assembled cells were characterized using a Solartron instrument (model 1260 with an electrochemical interface, Solartron, Houston, TX). Their charge and discharge capacities were measured by cycling them at constant currents between voltage limits, and interfacial impedances were determined from the ac impedance spectra measured at the 0.1–10^6 Hz frequency range.

Results and Discussion

The physical constants of the cell components are shown in Table I. The motivation for designing, selecting, and processing cell components was to enhance energy and power densities. The electrochemical performances of cells 2 and 4 are presented in Tables II and III, respectively. Table I shows that the physical parameters (thickness and weight) of the lithium anode remained constant. The physical parameters of the membrane laminates were varied. The thickness and weight were reduced by more than a factor of 2 if cells 1 and 4 were compared. The cathode formulation was also altered.

Table II. Electrochemical data on Li–O$_2$ cell 2.

T (C°)	D/C cycle	D/C current (mA)	D/C time (h)	Cell capacity (mAh)	R_{cell} (Ω)	Cell voltage excursion (V)
45	1D	0.20	0.85	0.17	4148	3.08–1.00
45	1C	0.20	0.51	0.10	816	3.23–4.50
57	4D	0.10	8.07	0.81	1585	3.09–1.00
57	4C	0.075	11.0	0.83	127	2.77–4.17
57	5D	0.20	4.18	0.84	1690	3.45–1.00
57	5C	0.075	12	0.90	185	2.98–4.00
57	6D	0.10	10.14	1.02	436	3.66–1.00
57	6C	0.075	15	1.12	323	3.12–4.05
67	7D	0.20	10.76	2.15	927	3.62–1.00
67	7C	0.10	22	2.20	123	2.63–4.00
75	8D	0.20	24.4	4.88	249	3.44–1.00
75	8C	0.125	40	5	156	2.35–4.04
85	9D	0.20	63.05	12.61	97	3.56–1.00
85	9C	0.20	64.03	12.81	92	2.76–4.18
95	10D	0.30	21.77	6.53	126	3.55–1.00
95	10C	0.30	22	6.60	70	2.14–4.09
95	11D	0.20	56.72	11.34	151	3.53–1.00
95	11C	0.30	38	11.40	107	2.80–4.21
105	12D	0.20	50.2	10.04	288	3.50–1.00
105	12C	0.30	34	10.20	155	2.55–4.15

Notations: T is the temperature, D is the discharge, C is the charge, and R_{cell} is the cell resistance in the D/C state.

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Table III. Electrochemical data on cell 4 during multiple charge/discharge at 75 and 85°C. The duration of each charge/discharge is 2 h.
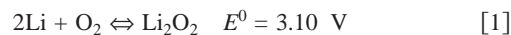
D	T (°C)	V_D range (V)	i_D (mA)	Cell capacity (mAh)	C	T (°C)	V_C range (V)	i_C (mA)	Cell capacity (mAh)
1	75	3.32–2.82	0.05	0.1	1	75	2.93–3.80	0.05	0.1
2	75	3.58–3.11	0.05	0.1	2	75	3.47–3.83	0.05	0.1
3	75	3.56–3.03	0.05	0.1	3	75	3.44–3.85	0.05	0.1
4	75	3.54–3.03	0.05	0.1	4	75	3.42–3.85	0.05	0.1
5	75	3.53–3.01	0.05	0.1	5	75	3.41–3.86	0.05	0.1
6	75	3.59–2.63	0.1	0.2	6	75	2.94–4.10	0.1	0.2
7	75	3.68–2.63	0.1	0.2	7	75	3.43–4.19	0.1	0.2
8	75	3.56–2.61	0.1	0.2	8	75	3.37–4.22	0.1	0.2
9	75	3.53–2.60	0.1	0.2	9	75	3.34–4.23	0.1	0.2
10	75	3.50–2.60	0.1	0.2	10	75	3.32–4.23	0.1	0.2
11	75	3.62–2.48	0.1	0.2	11	75	3.38–4.18	0.1	0.2
12	75	3.50–2.50	0.1	0.2	12	75	3.32–4.24	0.1	0.2
13	75	3.47–2.45	0.1	0.2	13	75	3.30–4.27	0.1	0.2
14	75	3.46–2.40	0.1	0.2	14	75	3.30–4.30	0.1	0.2
15	75	3.45–2.24	0.1	0.2	15	75	3.28–4.31	0.1	0.2
16	85	3.35–2.66	0.1	0.2	16	85	3.24–4.05	0.1	0.2
17	85	3.39–2.68	0.1	0.2	17	85	3.28–4.05	0.1	0.2
18	85	3.43–2.68	0.1	0.2	18	85	3.30–4.05	0.1	0.2
19	85	3.44–2.67	0.1	0.2	19	85	3.31–4.06	0.1	0.2
20	85	3.44–2.67	0.1	0.2	20	85	3.31–4.07	0.1	0.2
21	85	3.45–2.66	0.1	0.2	21	85	3.10–4.08	0.1	0.2
22	85	3.29–2.65	0.1	0.2	22	85	3.20–4.07	0.1	0.2
23	85	3.37–2.63	0.1	0.2	23	85	3.26–4.08	0.1	0.2
24	85	3.40–2.62	0.1	0.2	24	85	3.28–4.09	0.1	0.2
25	85	3.42–2.61	0.1	0.2	25	85	3.29–4.10	0.1	0.2
26	85	3.64–2.60	0.1	0.2	26	85	3.44–4.13	0.1	0.2
27	85	3.53–2.60	0.1	0.2	27	85	3.37–4.15	0.1	0.2
28	85	3.46–2.57	0.1	0.2	28	85	3.33–4.17	0.1	0.2
29	85	3.45–2.56	0.1	0.2	29	85	3.31–4.18	0.1	0.2
30	85	3.43–2.51	0.1	0.2	30	85	3.30–4.20	0.1	0.2

Notations: T is the temperature, D is the discharge, C is the charge, V_D is the discharge voltage excursion (V), V_C is the charge voltage excursion (V), i_D is the discharge current (mA), and i_C is the charge current (mA).
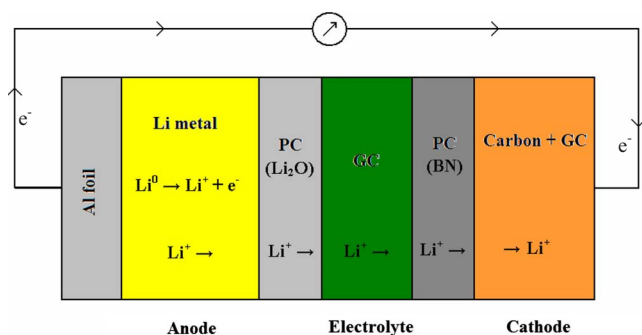
A key component of the solid-state Li–O_2 battery is the highly Li-ion conducting GC electrolyte synthesized from the LAGP system with superionic stoichiometry. An earlier investigation[11] showed that it has an ionic conductivity of about 10^{-2} S cm^{-1} at 30°C. A schematic of the cell configuration (Fig. 1) shows that a lithium anode and a carbon-based cathode are separated by an electrolyte laminate comprised of the GC membrane and two PC membranes. A PC membrane of PEO:LiBETI (8.5:1)–(1 wt % Li$_2$O) electrochemically coupled the lithium anode and the GC membrane except for cell 1. In cell 1, both the PC membranes of the cell were PEO:Li-

Figure 1. (Color online) A schematic of the Li–O_2 cell and its component materials are shown. The electrolyte laminate is composed of PC(Li$_2$O), GC, and PC(BN) membranes. The PC(Li$_2$O/BN) couples with lithium, which is protected by aluminum foil and used as a current collector. The current collector on the cathode side is a metal cell casing not shown in the schematic.

BETI (8.5:1)–(1 wt % BN). The cathode and the GC membrane in all cells were coupled by a PC membrane of PEO:LiBETI (8.5:1)–(1 wt % BN) stoichiometry. The details on the development of the PC membranes can be found elsewhere.[12,14,15] The PC membranes reduced the impedance of the cell, enhanced the charge transfer (Li^0 → Li$^+$ + e$^-$) at the anode, and electrochemically coupled the cathode to the GC membrane. It was also determined that Al foil covered the lithium surface effectively, protected the lithium, and provided stable cell impedance.[15]

The OCV of the Li–O_2 cell was measured as a function of state-of-charge/discharge, oxygen pressure on the cathode, and temperature. In the oxygen atmosphere, the OCV varies from 2.6 to 3.6 V depending on the state-of-charge/discharge. The OCV tends to increase after discharge and to attain equilibrium OCV of about 3.1 V. The charged state of the cell yields an OCV of around 3.6 V, which has a tendency to decrease and approach equilibrium OCV of about 3.1 V. These OCV variations could be explained on the basis of slow diffusion effects, which are perhaps a characteristic of solid-state cells. The equilibrium OCV of 3.1 V suggests that the electrochemical reaction in all probability proceeds according to Eq. 1 with Li$_2$O$_2$ as the reaction product
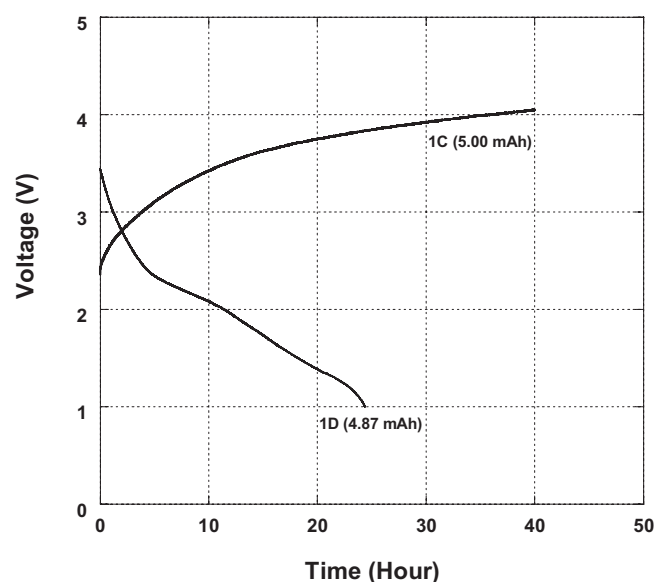
$$2Li + O_2 \Leftrightarrow Li_2O_2 \quad E^0 = 3.10 \text{ V} \quad [1]$$

The finding is consistent with the OCV of the plastic Li–O_2 cell reported by Abraham and Jiang[4] and the rechargeable Li–O_2 cell investigated by Ogasawara et al.[9] According to Eq. 1, the cells can be charged in low oxygen pressure or even in vacuum because the charging produces oxygen. It is only during the discharge cycle that oxygen is consumed to carry out the electrochemical reaction. Very
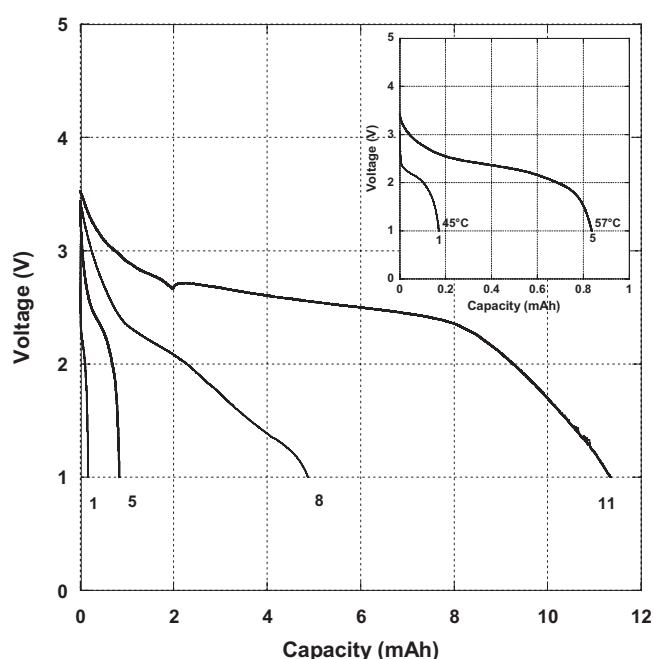
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Figure 2. The first discharge and charge curves of cell 2 show a continuous voltage variation in the range of 4.05–1.0 V. During discharge, the voltage shows a rapid decline as a function of capacity in the early stages of discharge. Subsequently, the slope of the discharge voltage is reduced. Toward the end of the discharge, the decline in voltage is prominent. The charge curve shows a relatively rapid increase in voltage during the early stage.

Figure 3. Discharge voltage shown as a function of capacity and temperature with a constant discharge current density of 0.1 mA/cm^2. Curves 1, 5, 8, and 11 correspond to 45, 57, 75, and 95°C, respectively. The discharge capacity shows a major enhancement as the temperature is increased to 95°C. The general shape of the discharge curves is similar. As can be expected from the conductivity data, the ohmic contribution at 95°C is reduced, leading to a major enhancement in the discharge capacity.

little discharge capacity was obtained in the absence of oxygen. The discharge capacity was also determined to be dependent upon the oxygen partial pressure.

The Li–O$_2$ cells were discharged in the temperature range of 30–105°C. The temperature-dependent discharge current density ranged from 0.025 to 0.25 mA/cm^2. In general, a lower discharge rate was used at temperatures below 75°C. Similarly, the charge rate varied from 0.025 to 0.15 mA/cm^2; again, it depended upon the temperature. The discharge and charge profiles of cell 2 at 75°C are shown in Fig. 2. The discharge and charge current density were 0.1 and 0.0625 mA/cm^2, respectively. The discharge voltage ranged from 3.5 to 1.0 V, whereas the charge voltage varied from 2.3 to 4.05 V. The discharge and charge capacities were 4.87 and 5.0 mAh, respectively. The shapes of the charge and discharge curves in Fig. 2 are different because they correspond to different rates (discharge/charge) and electrochemical reactions. During the discharge, an electrochemical reaction, such as that shown in Eq. 1 with Li$_2$O$_2$ as the reaction product, is carried out in the cathode. During the charge, lithium is plated onto the metallic lithium surface. These reactions are very different from the electrochemical reactions taking place in commercially available lithium ion batteries, which often show symmetrical discharge and charge curves.
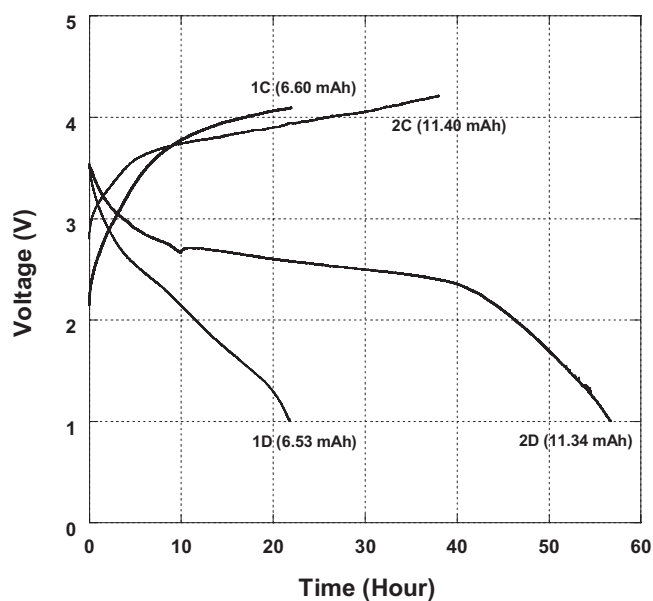
The dependency of the discharge capacity from 45 to 95°C with a constant discharge current density of 0.1 mA/cm^2 is shown in Fig. 3. The cell exhibited superior performance above 75°C, consistent with the conductivity of the electrolyte, which has a value of >100 mS cm^{-1} above 75°C. The inset in Fig. 3 shows the discharge curves at 45 and 57°C on an expanded scale. A small dip in the discharge curve of the cell at 95°C and at around 2 mAh capacity is attributed to either a flaw in the cell or an instrumental error. The observation has not been reproduced. Figure 3 illustrates that the temperature is a very important factor in determining the achievable capacity of the cell.

Figure 4 depicts the discharge curves of cell 2 at current densities of 0.10 and 0.15 mA/cm^2 at 95°C. The charge current density was maintained constant at 0.15 mA/cm^2 for both the cycles while maintaining the temperature at 95°C. By lowering the current density from 0.15 to 0.10 mA/cm^2, the cell capacity is improved by over 70%. The current density is another factor that determines the

cell capacity. The charge–discharge reversibility is excellent. This observation is very promising compared to the results reported by prior efforts.[4,9] Prior efforts reported a large hysteresis between the

Figure 4. The first two discharge and charge cycles of a Li–O$_2$ cell at 95°C as a function of capacity with discharge current densities of 0.1 and 0.15 mA/cm^2 are shown in Fig. 4. The charge current density in both cycles was 0.15 mA/cm^2. The discharge curves 1D and 2D correspond to capacities of 6.53 and 11.34 mAh, respectively. The charge curves 1C and 2C show charged capacities of 6.60 and 11.4 mAh, respectively.

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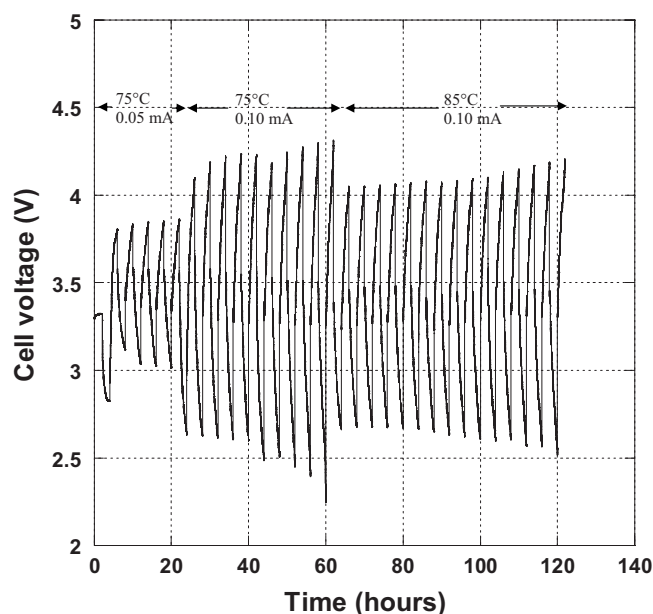
Figure 5. Rechargeability of Li–O$_2$ cell 4 as a function of charge/discharge current and temperature for a constant time (2 h) of charge and discharge. The first five cycles are at 75°C with a discharge/charge current of 0.05 mA. The second 10 cycles are at 75°C with the discharge/charge current of 0.1 mA. The last 15 cycles at 85°C correspond to the discharge/charge current of 0.1 mA. The cell was evaluated in an oxygen atmosphere, discharged down to about 2.5 V, and charged with the same current (as the discharge current) of up to about 4.3 V. The abscissa of the figure represents the cumulative time for both the discharge and charge cycles.

charge and discharge voltages. Also, in these studies, a poor rechargeability was observed without a transition-metal catalyst in the oxygen/air cathode.

The reversible behavior of the cell is attributed to the moderate operation temperature facilitated by the highly ion conducting solid electrolyte and possible catalysis of oxygen reduction by the LAGP material in the cathode. The major constituent of the cathode is the LAGP material, and the cathode does not contain a catalyst. Yet, the cathode capacity is high. The observation has led us to believe that the LAGP material with carbon catalyzes the reduction of oxygen.

Over two dozen lithium–oxygen cells have been fabricated and characterized in a span of about a year in our laboratory. The physical constants and electrochemical performances of four representative Li–O$_2$ cells are presented in Tables I-III. The cycling stability of the cells at isothermal conditions is shown in Tables II and III. The cycling of cell 4 at different discharge temperatures and currents is shown in Fig. 5. This cell was discharged and charged five times at 75°C with a constant current of 0.05 mA. The data exhibit a desirable rechargeability. Subsequently, discharge and charge currents were increased to 0.1 mA. At this discharge/charge current, the corresponding voltage range widened implying an increased loss and a cycling-related degradation within the cell. The discharge/charge voltage range was suppressed by increasing the cell operating temperature to 85°C. Cell 4 was cycled 30 times at 75 and 85°C. It remained functional for evaluation at higher temperatures of 95 and 105°C. The cell was subjected to additional five cycles each at 95 and 105°C (not shown in the figure).

A rechargeable Li–O$_2$ cell integrates the positive attributes of a fuel cell and a rechargeable battery. The Li–O$_2$ and H$_2$–O$_2$ cells provide identical functions. However, there are a few major advantages associated with a Li–O$_2$ cell. In a Li–O$_2$ cell, solid metallic lithium is the fuel that minimizes the containment and transportation

issues. The solid lithium also functions as an anode, whereas in a H$_2$–O$_2$ cell, an expensive membrane electrode assembly based on precious metals (Pt, Pd, etc.) must be used to carry out the oxidation of hydrogen and the reduction of oxygen at lower temperatures. Furthermore, the OCV of the Li–O$_2$ cell is about 3.0 V, which is approximately three times the OCV of a H$_2$–O$_2$ cell. The higher OCV translates into three times more power per cell for an identical current density. The reactivity of lithium and the associated safety issues have been addressed in this investigation by using a totally solid-state, mostly inorganic electrolyte that isolates lithium from moisture and atmospheric gases and prevent dendritic shorts.

Armand and Tarascon[3] in their recent paper assessed the future of the Li–O$_2$ battery and identified issues such as "high energy density but poor energy efficiency and rate capability" and "rechargeability to be proven." The data presented in this paper address these issues, suggest their solutions, and provide directions for "building better batteries" in the future. At the current stage of the Li–O$_2$ cell development, the measured energy density is about 750 Wh/kg based on the weight of the cell, including an allowance for packaging.

Conclusions

In this paper, the fabrication, characterization, and performance of solid-state, rechargeable, and long cycle life Li–O$_2$ cells are presented and discussed. The cells consisted of a lithium metal anode, a membrane laminate fabricated from GC and PC materials, and a solid-state composite oxygen/air cathode prepared from carbon and GC powder. The cells demonstrated excellent thermal stability and rechargeability in the 30–105°C temperature range. A cell was subjected to 40 discharge/charge cycles in the 30–105°C temperature range. The cells exhibited excellent reversibility.

The cathode formulations and processing, charge and discharge rates, and thickness of cell components were varied. These cells have produced extremely reproducible results, and the effort to improve their electrochemical performance continues. The results have led us to believe that the fabrication of a totally solid-state, safe, rechargeable, and commercially viable lithium–air battery with high energy and power densities is well within the reach.

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